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DETERMINATION OF POTENTIAL SOURCES OF AREA ATMOSPHERIC RADIO-ACTIVE CONTAMINATION

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Reactor Technology Division

DETERMINATION OF POTENTIAL SOURCES OF AREA ATMOSPHERIC RADIO-ACTIVE CONTAMINATION

C. P. Coughlen

DATE ISSUED:

JUN 8 1950

OAK RIDGE NATIONAL LABORATORY
Operated by
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1.0 INTRODUCTION

In May, 1948 it was learned that personnel at Hanford were somewhat concerned about airborne particles having high specific activities produced in the process of carrying out certain phases of their operations. In view of this it appeared desirable to study quite thoroughly the possibilities of hazards from such airborne activities at the Oak Ridge National Laboratory.

The results of such studies, revealing the presence within the plant area, of radioactive particles, seemed to emphasize the position that there might exist a hazardous condition.

Subsequent investigations by the Health Physics Division proved that radioactive particles, ranging in size from less than one micron to several hundred micron diameter, existed in an appreciable concentration and distribution throughout the plant area. In the absence of biological evidence that the indicated particle concentrations did not constitute a serious health hazard, it was concluded that the problem was sufficiently serious to warrant the expenditure of considerable effort in evaluating and reducing it.

This report is concerned with the results of the efforts expended by the Technical Division toward determining the activity output levels of various potential sources. Two sources proved to be major were evaluated after remedial measures were taken.

The results are reported as area contamination potentials since only the total quantity of active particulate material that issued from each location was determined. No estimate of the fraction of that material which settled within the boundaries of the area is attempted.

The reader is referred to the reports "ORNL-283 (Secret)" by J. S. Cheka and H. J. McAlduff, Jr., and "ORNL-267 (Secret)", Technical Division Quarterly for a more comprehensive survey of the discovery of the problem and the solutions effected.

2.0 TEST EQUIPMENT AND TEST PROCEDURES

Sampling equipment consisted simply of CWS filter papers mounted behind Aerotec cyclones, (optional), the sample stream flows being

[redacted]

2.0 TEST EQUIPMENT AND TEST PROCEDURES (CONT'D)

effected by Root Conners type blowers or by air jets. Sampling lines projected into the fume lines and were designed to provide fume line conditions of flow at the intake, to provide smooth-wall flow conditions and to give sampling line velocities equal to or slightly greater than the respective fume line velocities. Where possible, sample points were located to provide sufficient straight-line flow upstream to aid in the taking of a representative sample. Filtered sample streams were discharged either to the same fume lines (downstream), to adjacent lines, or to ducts properly equipped for safe discharge.

The equipment was designed to remove particulate matter only, it being considered that active gaseous matter represented no particular hazard for contamination.

Samples were drawn from the lines through the sampling equipment, suitable main line flow and sampling flow measurements being made. The resulting papers and cyclone jar contents were counted in total or in part in the ion chamber. All results were reported on the uniform basis of millicuries of gamma active material of one MEV per photon and one photon per disintegration.

Sampling as much as was possible, was done across each step of each operation to permit the determination of the major contributing operations.

Figure 1 is a schematic sketch of the equipment used.

3.0 ACTIVE PARTICULATE DISCHARGE MEASURMENTS

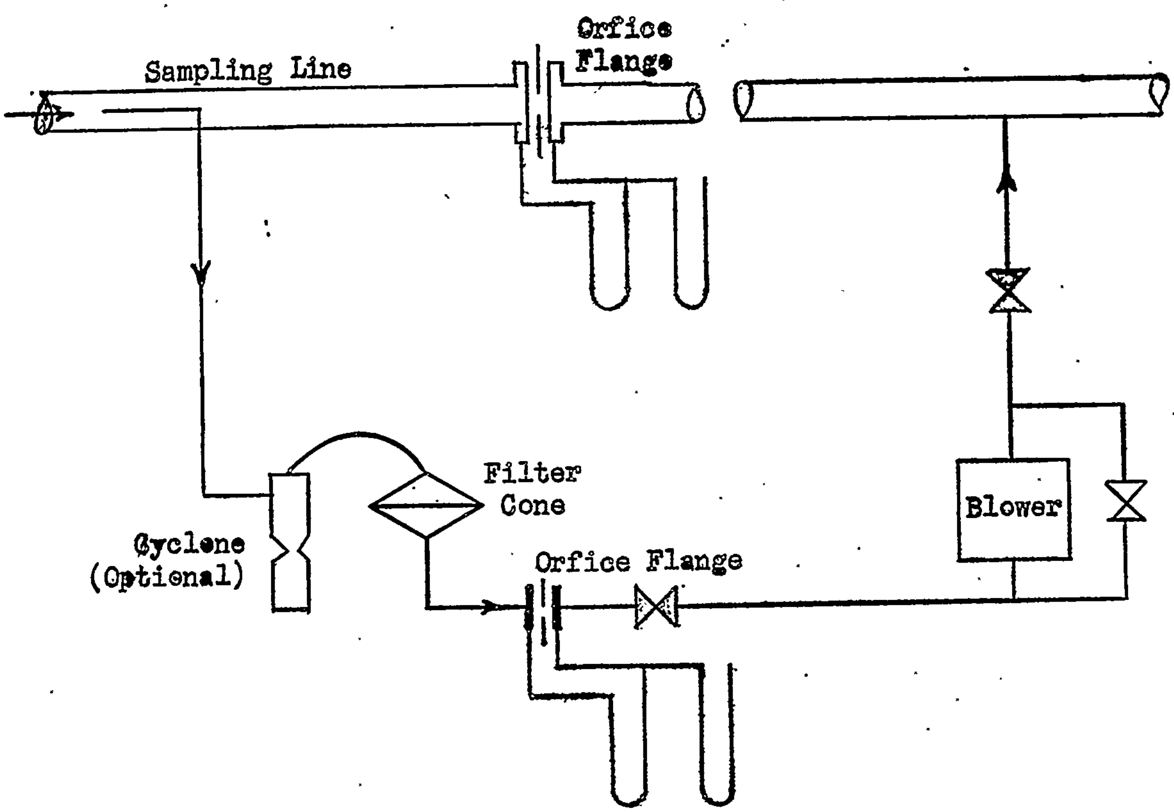
3.1 Particulate Discharges from RaLa Operation Fume Lines

Run #28 was sampled partially but will not be reported here. The detailed data resulting may be found in the report "Radiation Hazards Measurements for the Period November 26 to December 3", December 7, 1948 - ORNL Central Files No. 48-12-104. One item of pertinent information resulted. The Cell Ventilation line proved to be a major contributor.

Run #29 was sampled completely - and further, was sampled across each operation. Prior to sampling, a filter house (FG-50 backed by CWS paper) was installed in the Cell Ventilation line.

[redacted]

FIGURE I



SCHEMATIC LAYOUT - EQUIPMENT FOR SAMPLING ACTIVE PARTICULATES IN FUME LINES

3.1 Particulate Discharges from RaLa Operation Fume Lines (Cont'd)

Figures 2 to 5 show graphically the outputs of the Dissolver Off-Gas Line, the Vessel Off-Gas Line, the Cell Ventilation Line (before the filters), and the Cell Ventilation Line (after the filters). The curves in each figure represent the cumulative values, of all sampling periods, the decays of each specimen being considered. The bar graphs represent the individual contribution (at the time of removal of the sampling filters and cyclone jars) of each period.

Extensive work was invested in following the decays of each specimen, in the preparation of summary curves, and in the preparation of various comparisons. Little information of value, beyond that appearing on the charts, resulted.

3.2 Particulate Discharge from Iodine 135 Recovery Operations

Run #11, using X-10 slugs, was sampled completely. Figures 6 and 7 show graphically the outputs. The curves and graphs were developed similarly to those presented for RaLa operations.

Figure 8 shows the results obtained for the Cell Ventilation Line only during Run #12, a "double-header" in that two charges of slugs were used. The marked difference in activity output during the centrifuging operations (Run #11 vs. Run #12) may be explained by the inadvertent omission (due to an accident) of the washing of the cake by pipette decantation.

3.3 Particulate Discharge from Iodine 131 Recovery Operations

Runs #38 and 39, using X-10 slugs, were sampled completely. Figures 9 and 10 show graphically Run #38 data and Figures 11 and 12 are similar plots for Run #39 data. All values are quite low.

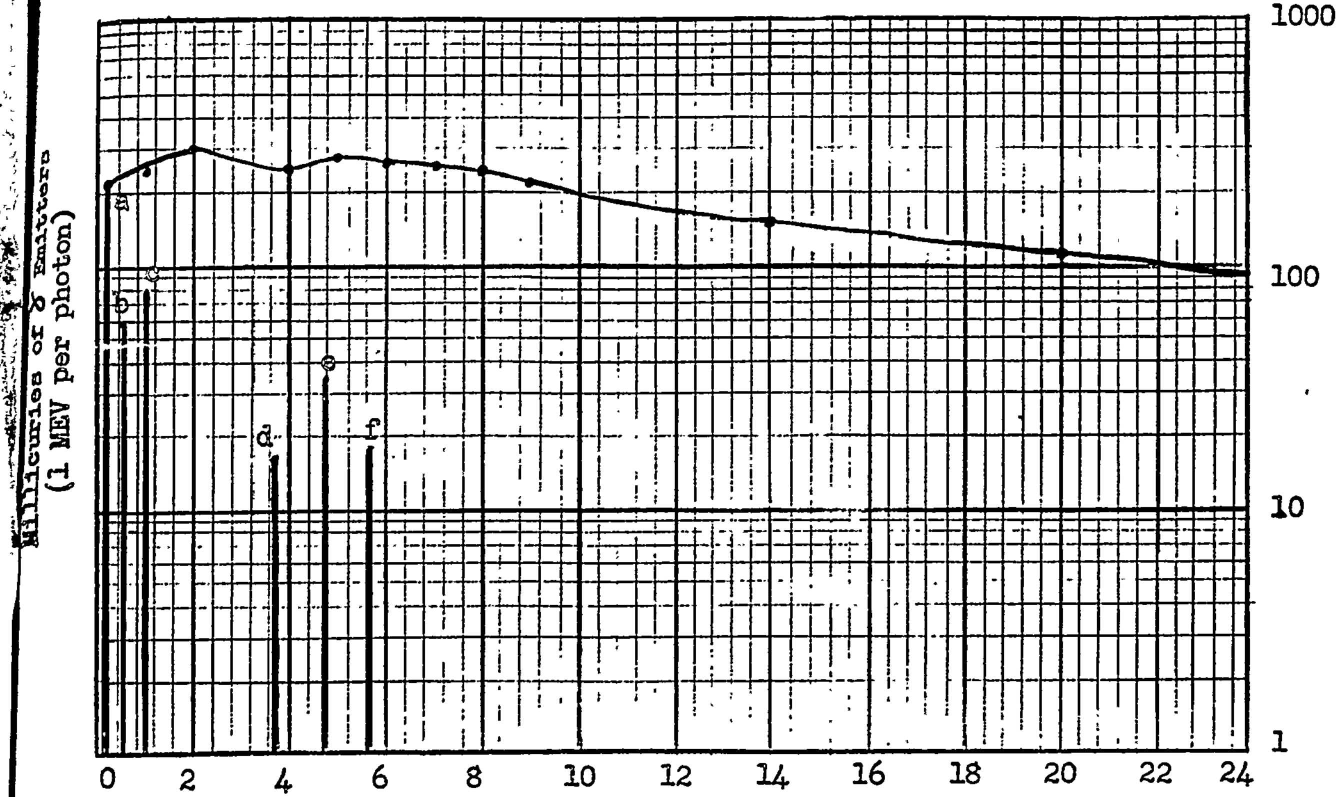
3.4 Particulate Discharge from Hot Pilot Plant Redox Operations

Two runs were sampled completely; one whose charge was made up of slugs 30% from Hanford (70% from ORNL) and another whose charge was 100% Hanford slugs. The dissolver off-gas, vessel off-gas and cell vent lines were sampled. Here the sampling was not coordinated with specific operations. Figures 13 to 15 show graphically the results on the 30% run while Figures 16 to 18 show the results obtained during the 100% Hanford run. (The 100% dissolver values were obtained by Bartholomew et al - M.I.T. Practice School).

[redacted]

FIGURE 2

Dwg.#8870



DAYS FROM START OF RUN

RALA #29 DISSOLVER OFF-GAS PARTICULATE OUTPUT

SAMPLING PERIOD LEGEND

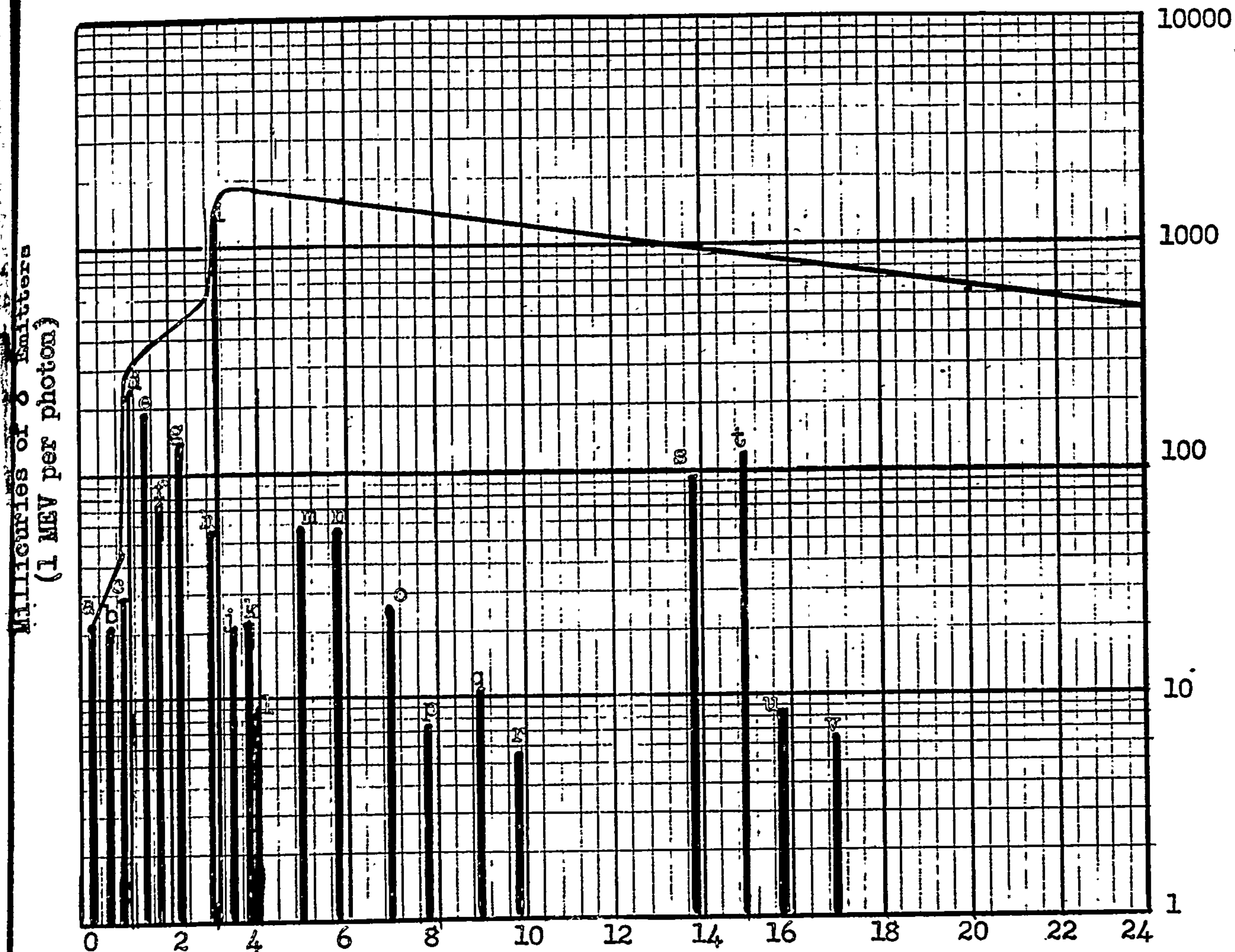
a - Jacket Removal and Dissolving
b - Down Period
c - Heel Dissolving - (Line down for 63 hours after this period)
d - Neutralization of 1/2 of Main Waste
e - Neutralization of Recovery Waste
f - Vessel Clean-out and Shutdown

[redacted]

[redacted]

FIGURE 3

Dwg #8871



DAYS FROM START OF RUN

RALA #29 VESSEL OFF-GAS PARTICULATE OUT PUT

SAMPLING PERIOD LEGEND

a - Digestion
b - Hot Settling
c - Cold Settling
d - Miscellaneous Operations
e - Heel Operations
f - Extraction Washes
g - Metathasis and Washes
h - Transfer to Cell B and Electrolysis
i - Transfer to B-6, Sampling and Volume Reduction
j - Transfer to Glassware and Glassware Operations
k - Product Evaporation
l - Waste Neutralization (1/2 of Wastes)
m - Recovery Cycle of Waste Settlings
n - Vessel Cleanout and Shutdown
o, p, q, r, s, t*, u, v - Shutdown Periods

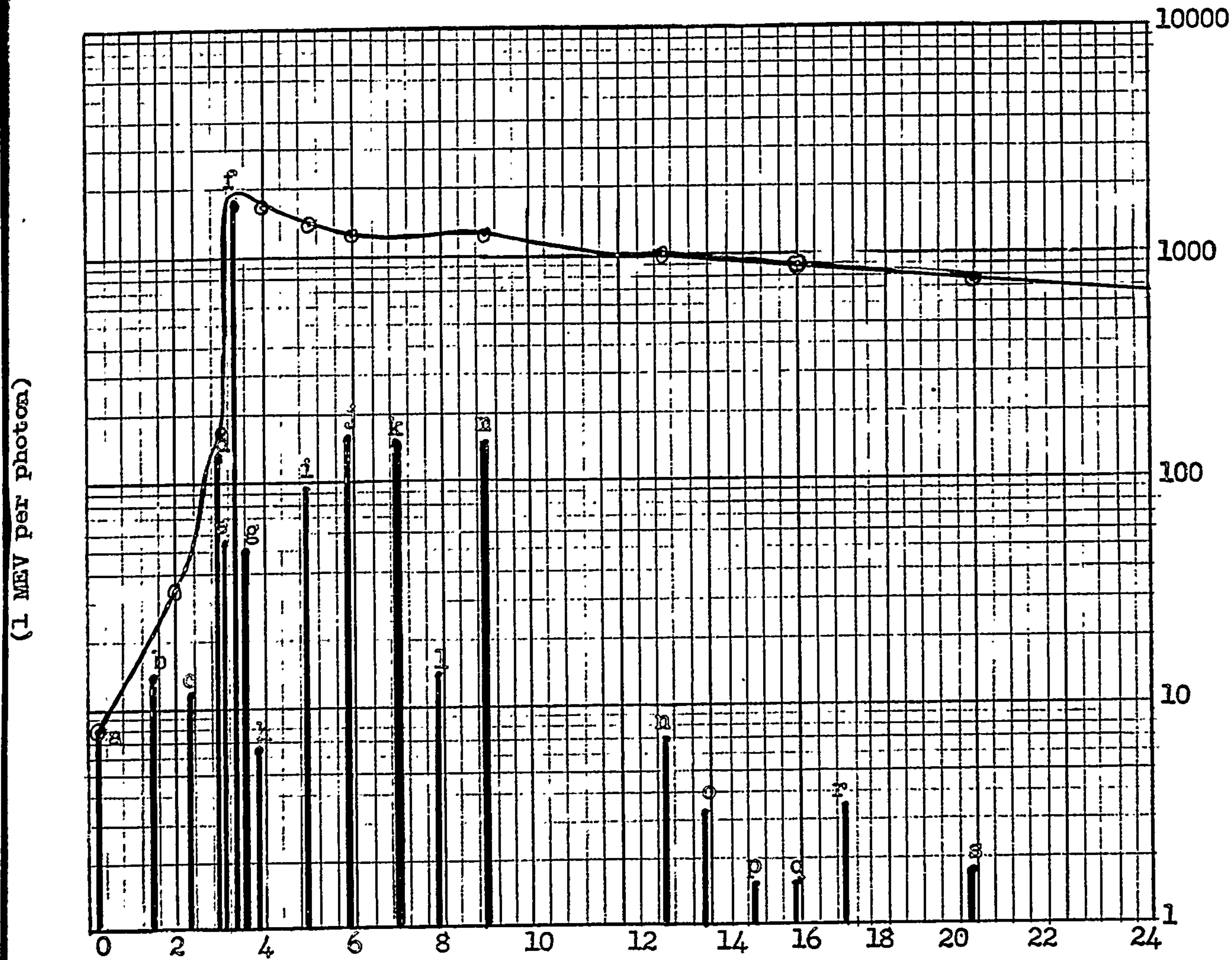
*- Period "t" probably includes line contamination for other operations.

[redacted]

[redacted]

FIGURE 4

Dwg.#8872



DAYS FROM START OF RUN

RALA #29 CELL VENTILATION - BEFORE FILTERS

SAMPLING PERIOD LEGEND

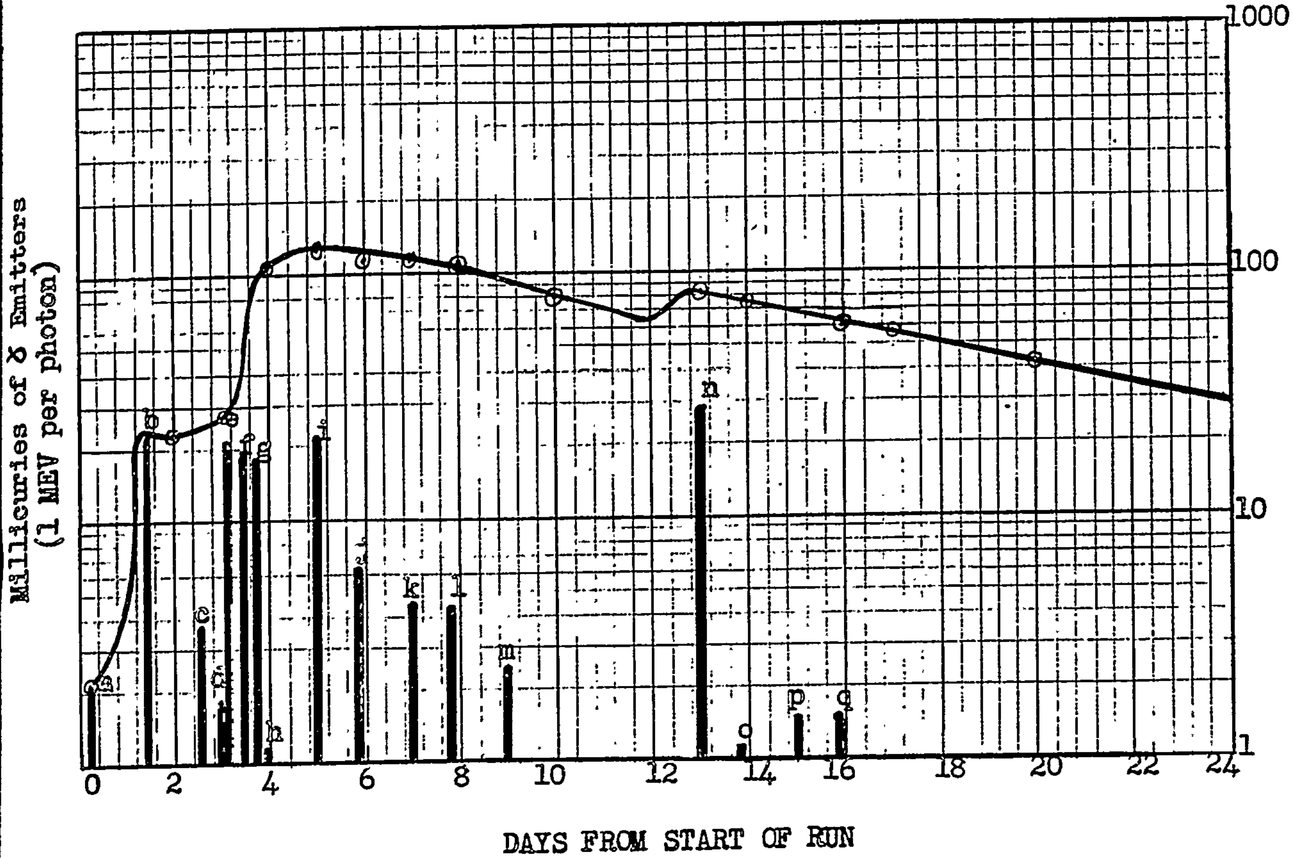
a - Coating Removal and Slug Dissolving
b - All other Cell-A Solution Operations
c - Extraction Washes; Metathasis and washes
d - Transfer to Cell B; Electrolysis
e - Transfer to B-6; Volume Reduction
f - Transfer to Glassware; Glassware Operations
g - Product Evaporation
h - Metathasis and Neutralization of Wastes
i - Remainder of Waste Cycle
j - Vessel Cleanout and Shutdown
k, l, m, n, o, p, q, r, s - Shutdown Periods

[redacted]

FIGURE 5

Dwg.#8873

[redacted]



RALA #29 CELL VENTILATION - AFTER FILTERS

<u>SAMPLING PERIOD LEGEND</u>

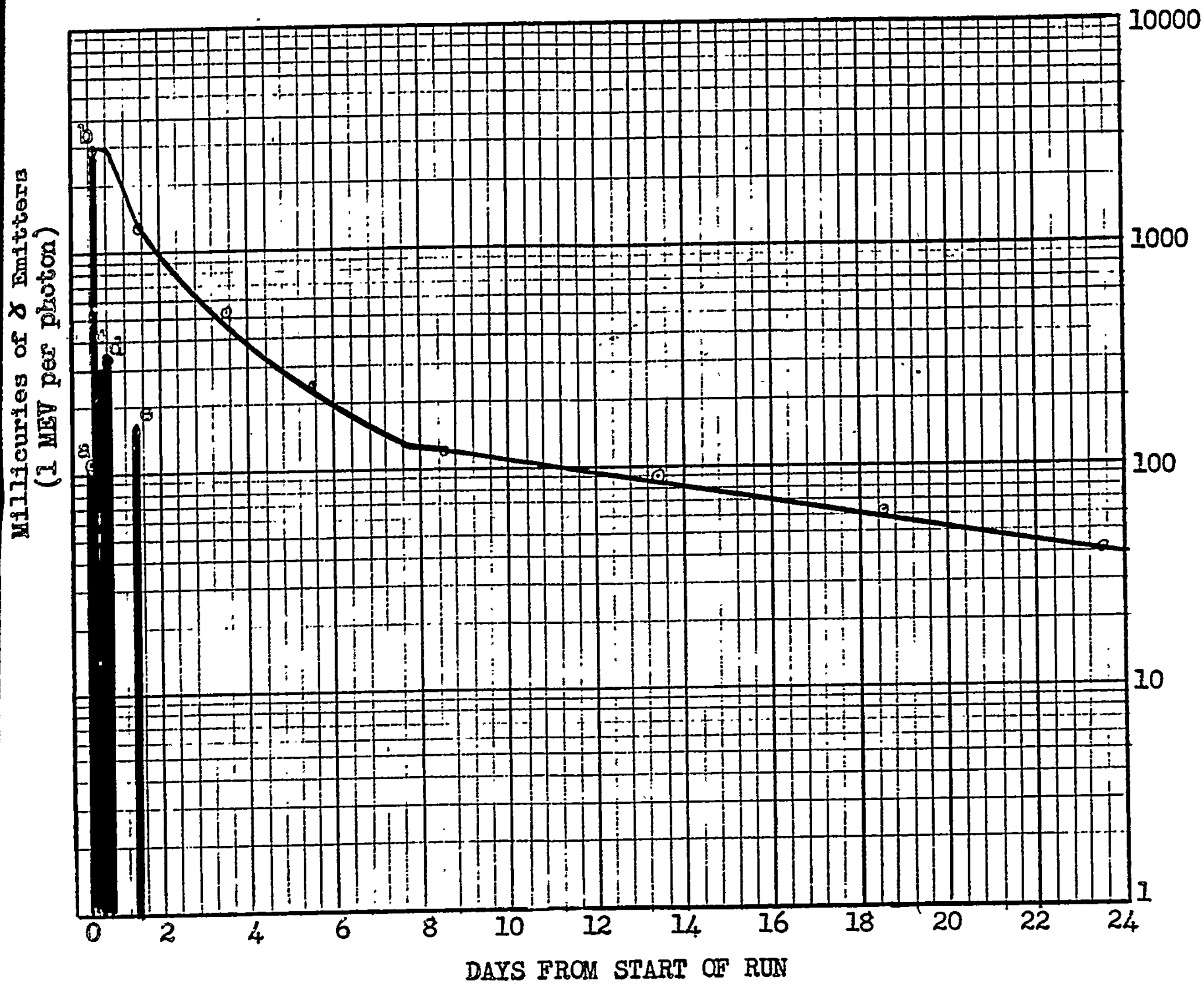
a - Coating Removal and Slug Dissolving
b - All other Cell - A Solution Operations
c - Extraction Washes; Metathasis and Washes
d - Transfer to Cell-B; Electrolysis
e - Transfer to B-6; Volume Reduction
f - Transfer to Glassware; Glassware Operations
g - Product Evaporation
h - Metathasis and Neutralization of Wastes
i - Remainder of Waste Cycle
j - Vessel Cleanout and Shutdown
k, l, m, n, o, p, q - Shutdown Periods

[redacted]

[redacted]

FIGURE 6

Dwg.#8874



I-135 RUN #11, VESSEL OFF-GAS LINE

SAMPLING PERIOD LEGEND

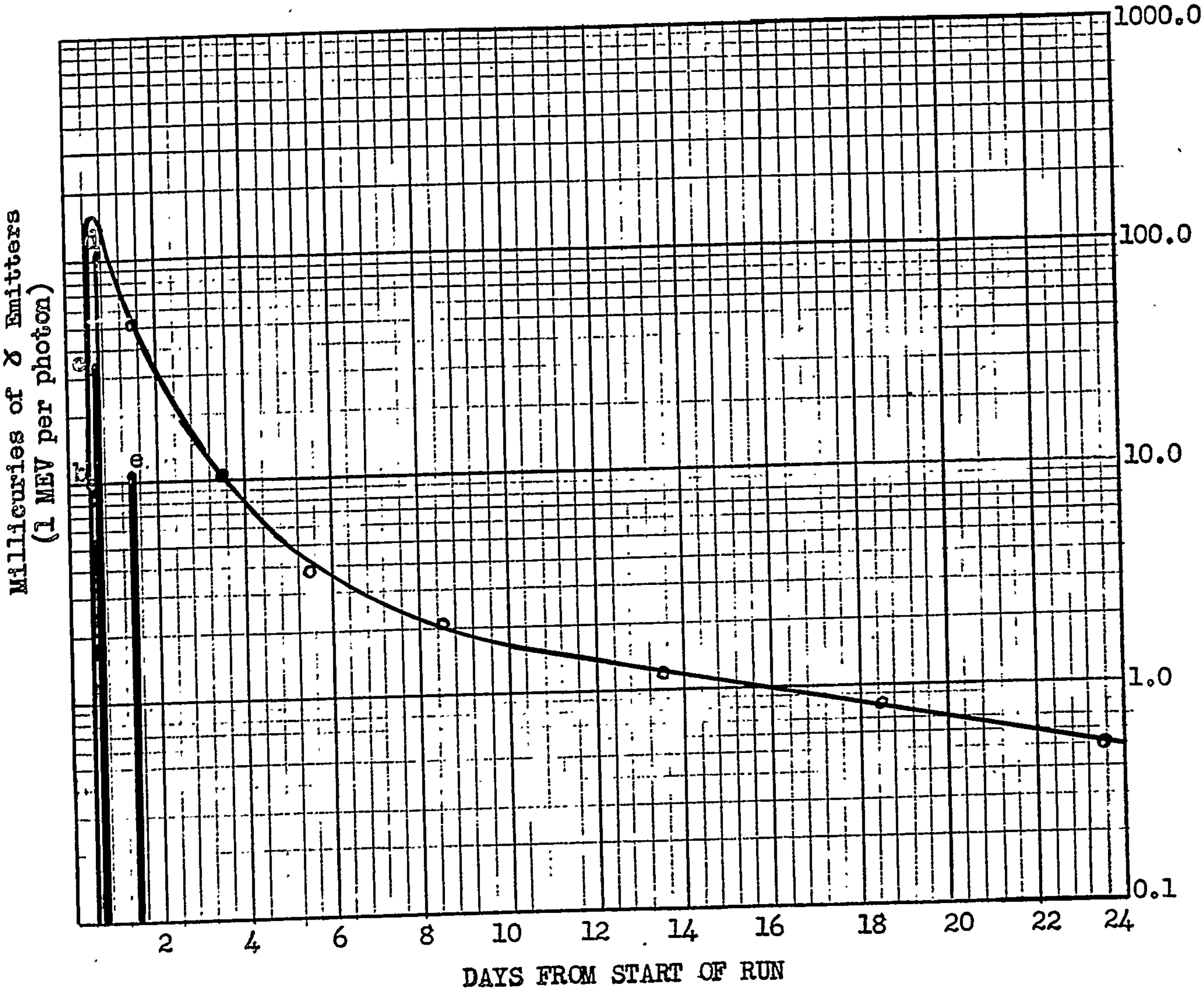
a - Slug Dissolving
b - Transfer, Oxidation and Distillation
c - Precipitation
d - Centrifuging
e - Waste Dumping, Dismantling and Clean-Up

[redacted]

Dwg #8875

[redacted]

FIGURE 7



I-135, RUN #11, CELL VENTILATION

<u>SAMPLING PERIOD LEGEND</u>

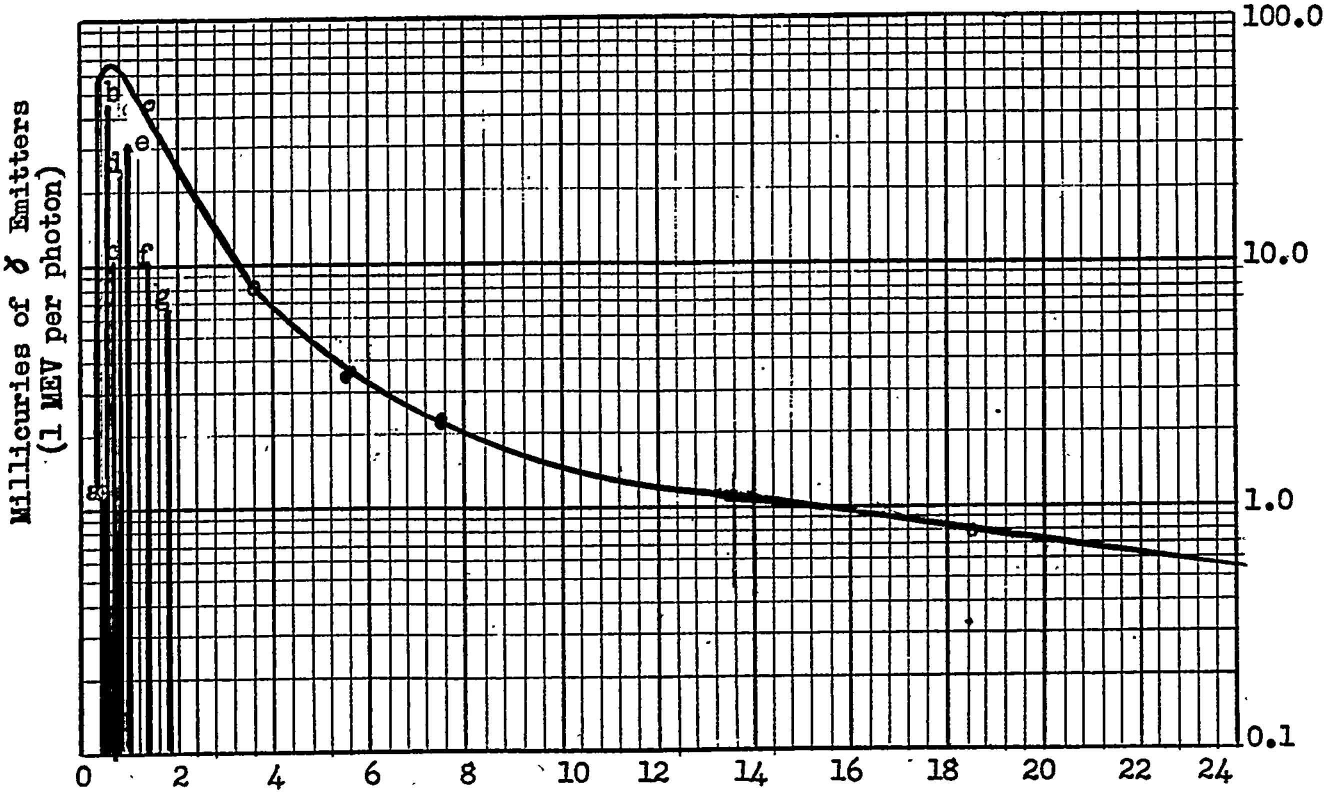
a - Slug Dissolving (< 0.1)
b - Transfer, Oxidation and Distillation
c - Precipitation
d - Centrifuging
e - Waste Dumping, Dismantling and Clean-Up

[redacted]

[redacted]

FIGURE 8

Dwg #8876



DAYS FROM START OF RUN

I-135, RUN #12, CELL VENTILATION

SAMPLING PERIOD LEGEND

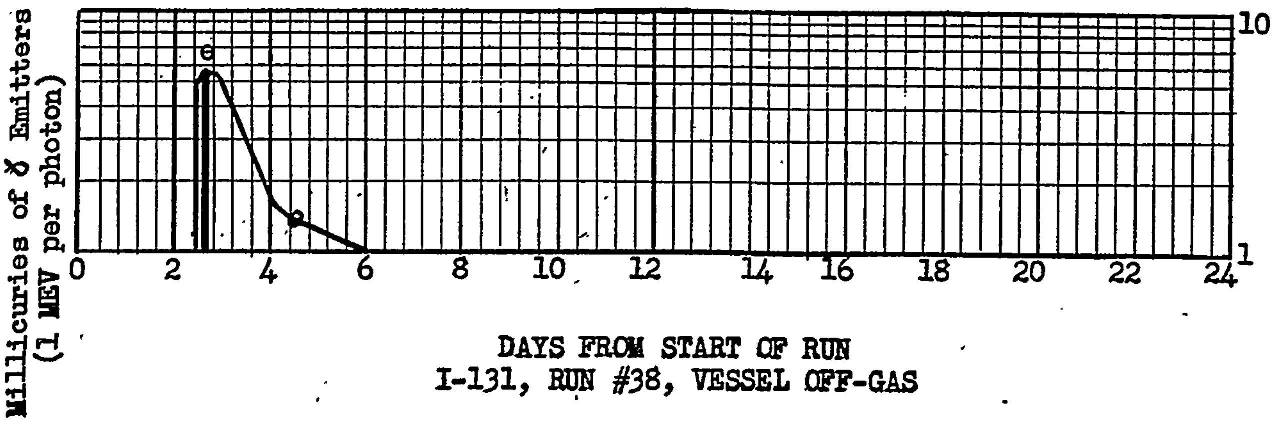
a - Dissolvings; First Transfer and First Oxidation
b - First Distillation; First metal Disposal
c - Second Transfer and Oxidation; Second I_2 Distillation
d - Precipitation and Cooling
e - Centrifuging
f - Dismantling and Clean-Up
g - Shut-Down

[redacted]

[redacted]

FIGURE 9

Dwg #8877



DAYS FROM START OF RUN
I-131, RUN #38, VESSEL OFF-GAS

SAMPLING PERIOD LEGEND

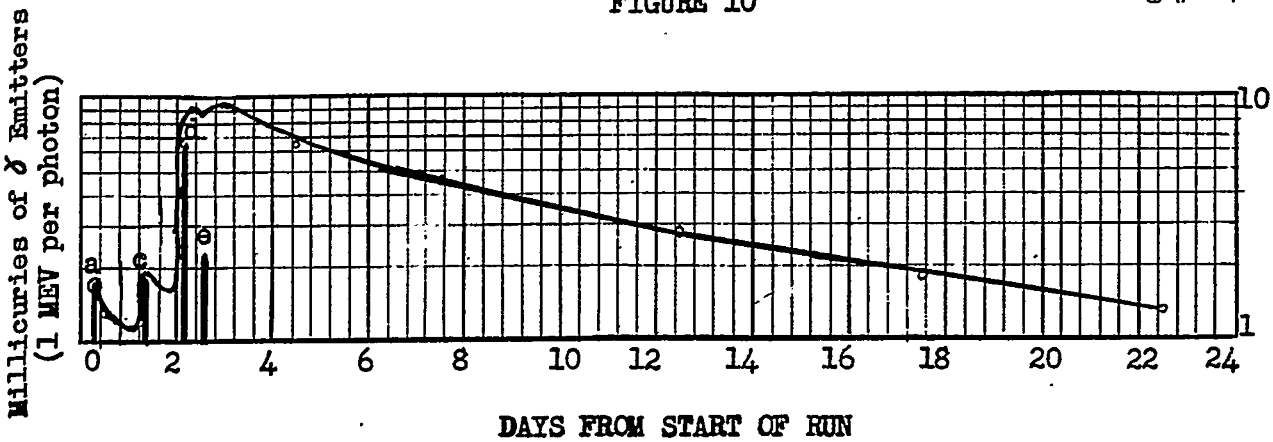
a - Slug Dissolving (< 1.0)
b - Steam Sparge (< 1.0)
c - Distillations; Initial Evaporation (< 1.0)
d - Glassware Operations (< 1.0)
e - Final Evaporation
f - Clean-Up and Shut-Down (< 1.0)

[redacted]

[redacted]

FIGURE 10

Dwg #8878



I-131, RUN #38, CELL VENTILATION

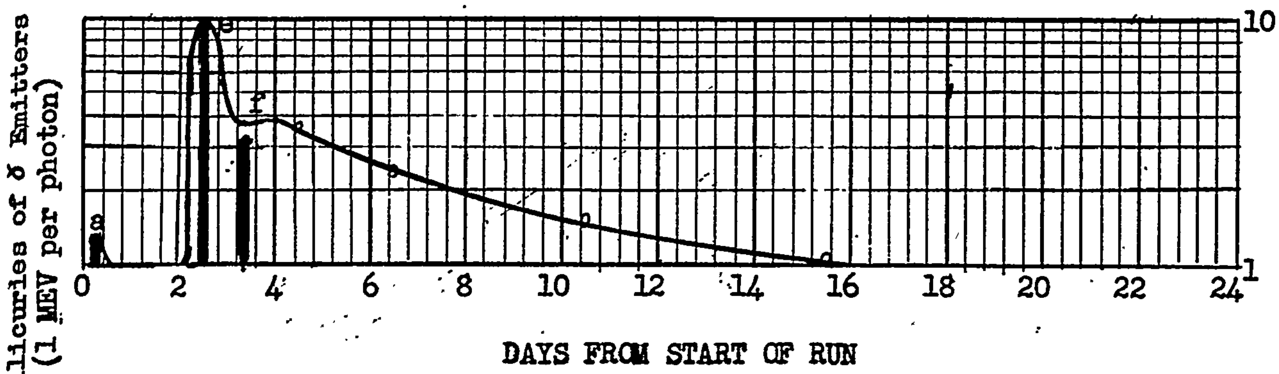
SAMPLING PERIOD LEGEND

a - Slug Dissolving
b - Steam Sparge (< 1.0)
c - Distillations; Initial Evaporation
d - Glassware Operations
e - Final Evaporation
f - Clean-Up and Shut-Down (< 1.0)

[redacted]

[redacted] Dwg.#8879

FIGURE 11



I-131, RUN #39, VESSEL OFF-GAS

SAMPLING PERIOD LEGEND

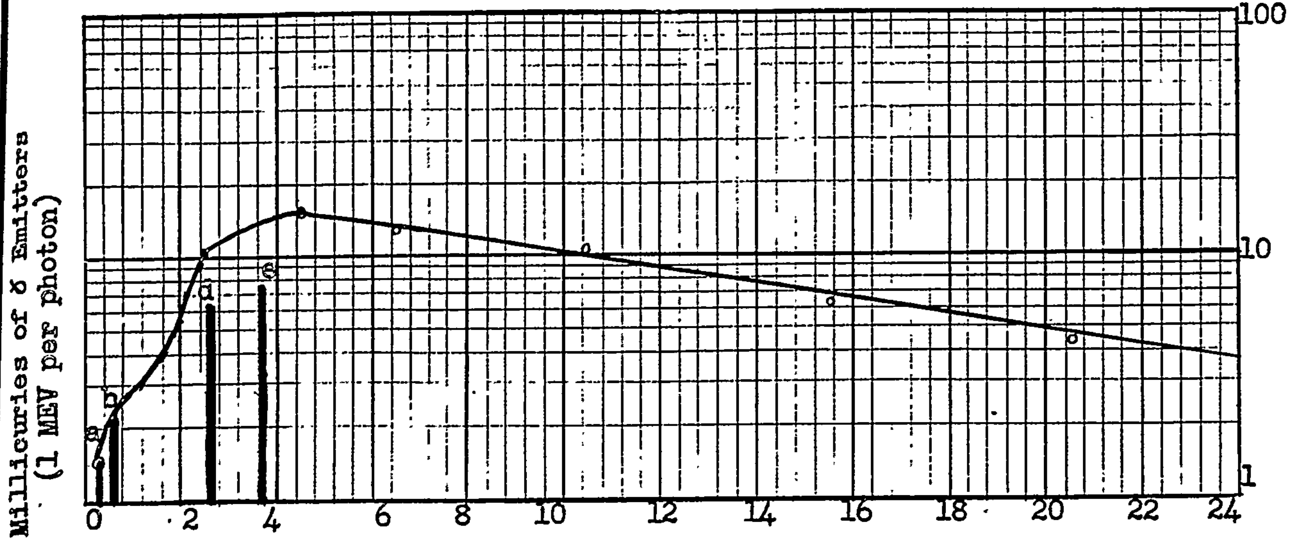
a - Slug Dissolving
b - Steam Sparge (< 1.0)
c - Distillation; Initial Evaporation (< 1.0)
d - Glassware Operations (< 1.0)
e - Final Evaporation
f - Clean-Up and Shut-Down

[redacted]

[redacted]

Dwg #8880

FIGURE 12



DAYS FROM START OF RUN
I-131, RUN #39, CELL VENTILATION

SAMPLING PERIOD LEGEND

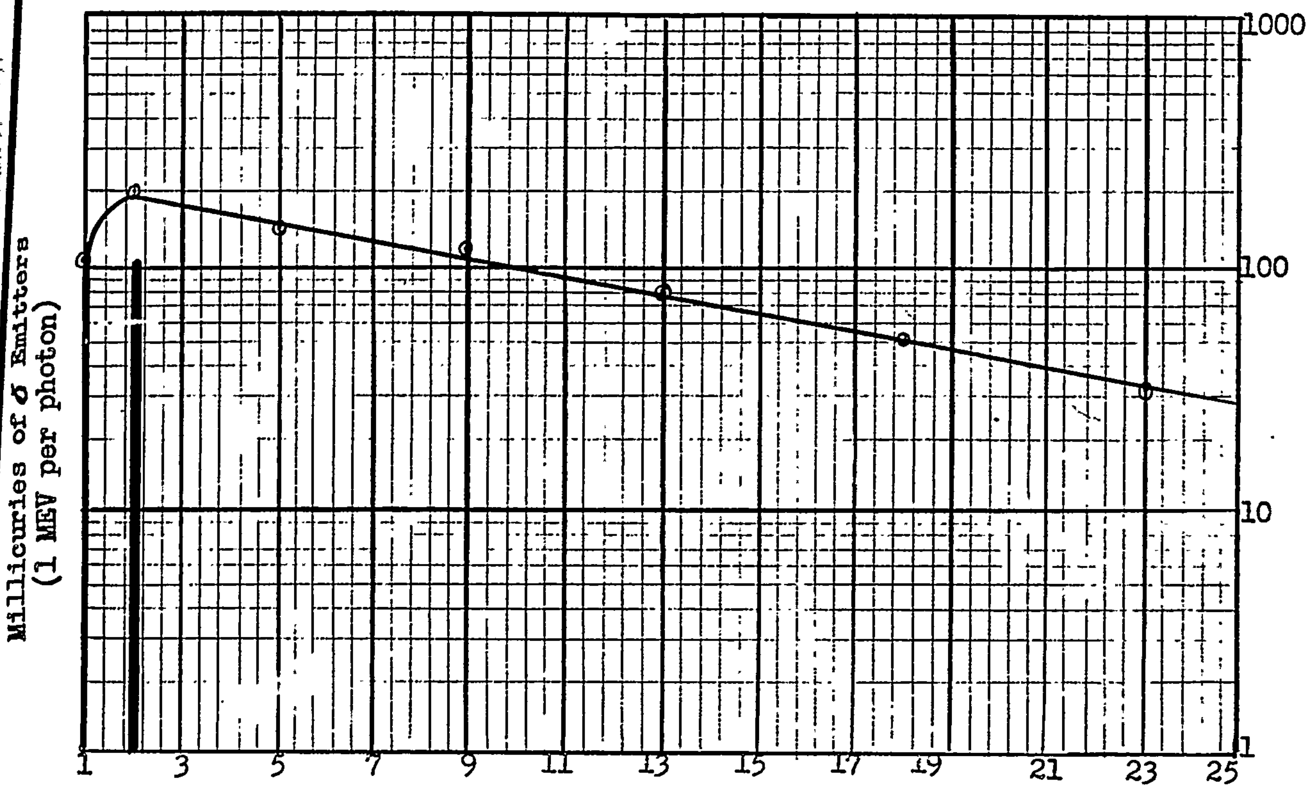
a - Slug Dissolving
b - Steam Sparge
c - Distillations; Initial Evaporation (< 1.0)
d - Glassware Operations
e - Final Evaporation
f - Clean-Up and Shut-Down (< 1.0)

[redacted]

[redacted]

FIGURE 13

Dwg.#8881



DAYS FROM START OF RUN

HOT PILOT PLANT DISSOLVER LINE - 30% HANFORD LEVEL

SAMPLING PERIOD LEGEND

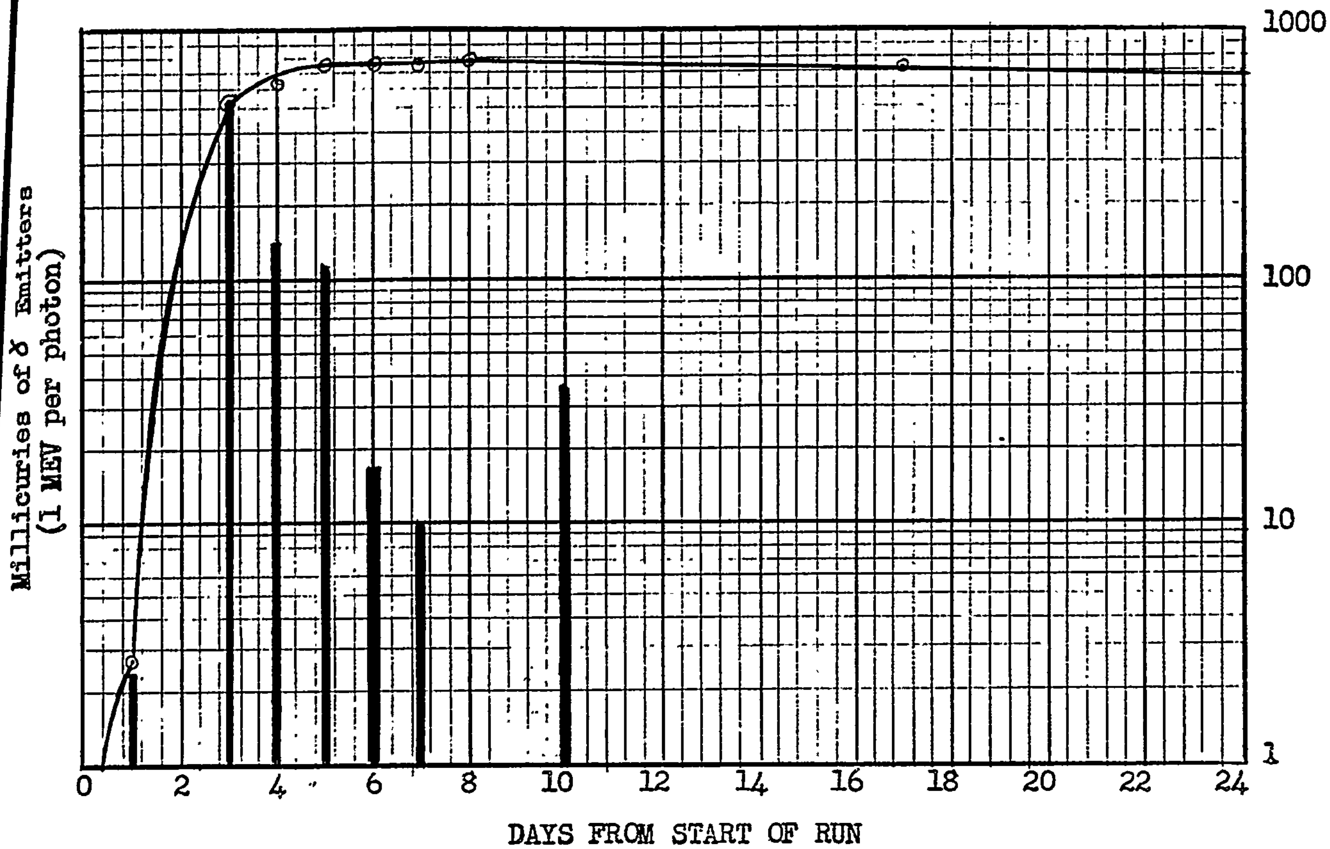
Sampling papers were removed periodically during one dissolving cycle.

[redacted]

[redacted]

FIGURE 14

Dwg #8882



HOT PILOT PLANT VESSEL OFF-GAS LINE - 30% HANFORD LEVEL

SAMPLING PERIOD LEGEND

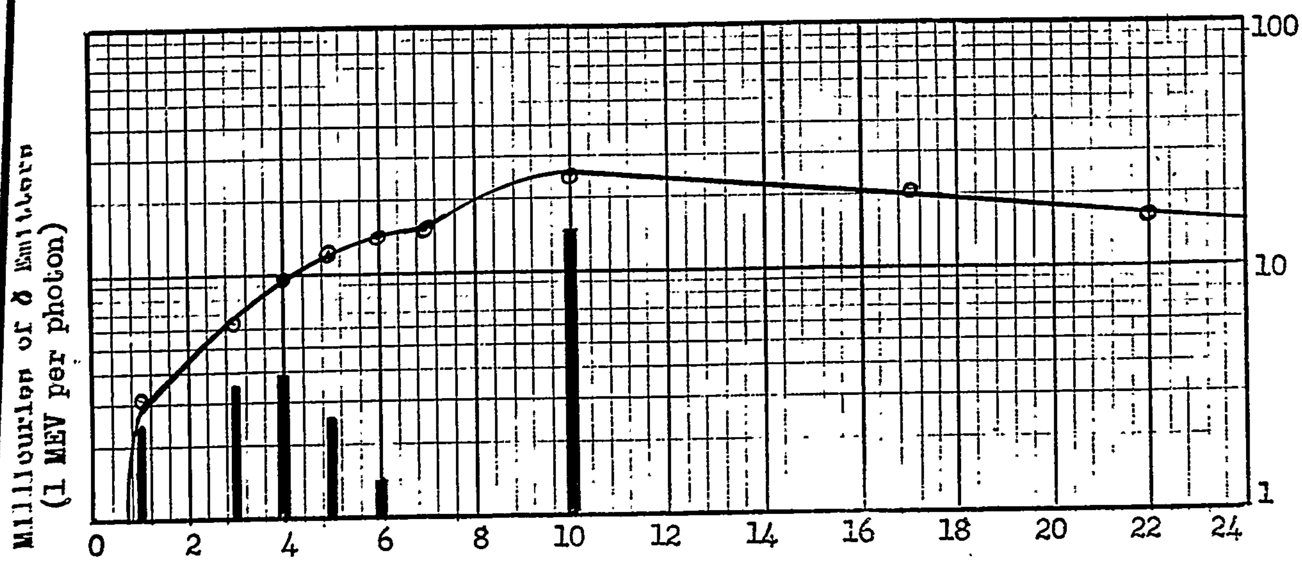
Sampling papers were removed periodically until the completion of the cycle.

[redacted]

[redacted]

FIGURE 15

Dwg #8883



DAYS FROM START OF RUN
HOT PILOT PLANT CELL VENTILATION - 30% HANFORD LEVEL

SAMPLING PERIOD LEGEND

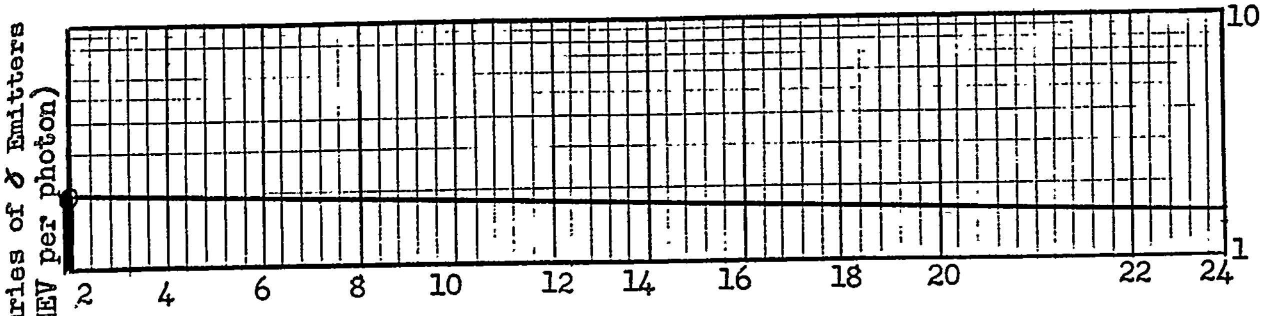
Sampling papers were removed periodically until the completion of the cycle.

[redacted]

FIGURE 16

[redacted]

Dwg.#8884



DAYS FROM START OF RUN

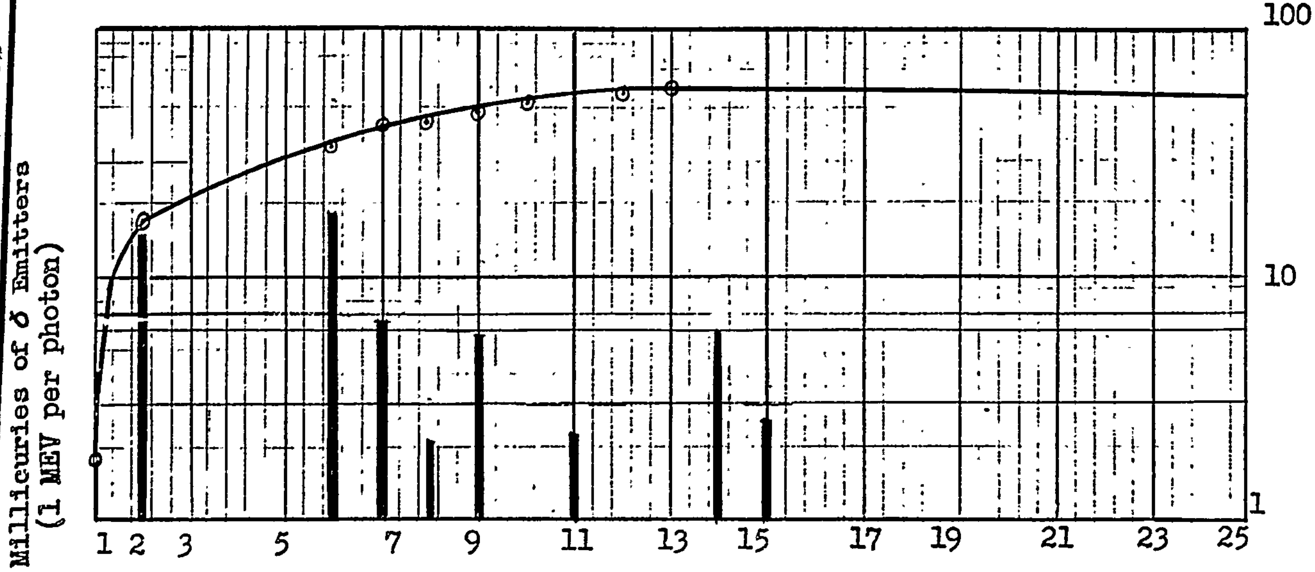
HOT PILOT PLANT DISSOLVER OFF-GAS LINE - 100% HANFORD LEVEL

SAMPLING PERIOD LEGEND

Sampling papers were removed periodically until the completion of the cycle.

[redacted]

[redacted] **FIGURE 17** Dwg #8885



DAYS FROM START OF RUN

HOT PILOT PLANT VESSEL OFF-GAS LINE - 100% HANFORD LEVEL

SAMPLING PERIOD LEGEND

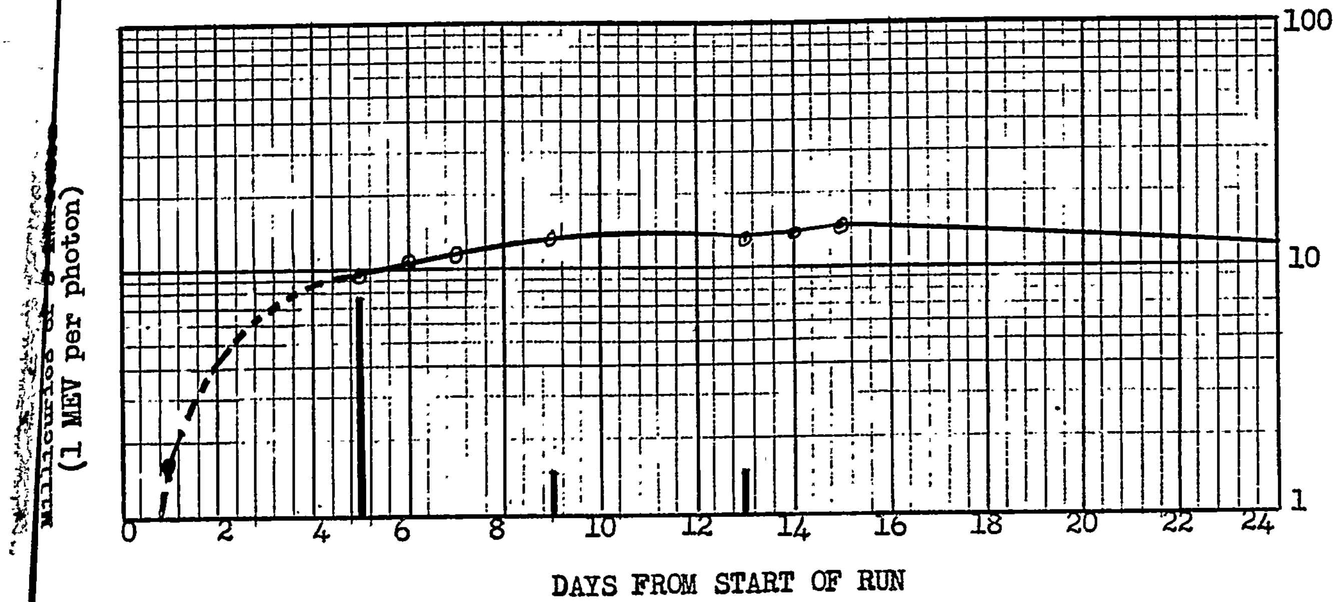
Sampling papers were removed periodically until the completion of the cycle.

[redacted]

[redacted]

FIGURE 18

Dwg.#8886



HOT PILOT PLANT CELL VENTILATION- 100% HANFORD LEVEL

SAMPLING PERIOD LEGEND

Sampling papers were removed periodically until the completion of the cycle.

[redacted]

[redacted]

3.4 Particulate Discharge from Hot Pilot Plant Redox Operations (Cont'd)

The relatively low activity output at the 100% Hanford level is attributed to the 120 + day aging the slugs received as compared to the 40 day aging the slugs received for the 30% run.

3.5 Particulate Discharge from Hot Hoods

Certain hoods were arbitrarily chosen for evaluation as being most likely to have particulates in their discharges. With hood sampling, a departure in technique is represented by the duration of sampling periods - from two to seven days. Also hoods were sampled two or three at a time since a single hood stack usually serves more than one hood. Sample pick-ups were made at the stack discharge points.

The following values are based on gamma-activity, assuming one MEV of gamma energy per disintegration.

TABLE I

Building 706-C

Hood Stack	Maximum Activity Level Mc/Hr.	Average Activity Level Mc/Hr.
Room I, Hoods 1, 2, and 3*	0.008	0.002
Rooms I and II, Hoods 4, 5	0.003(5)	0.001
Room V, Hood 8	0.0009	0.0003
Room IV, Hood 7	0.00009	0.00005
Room V, Hood 8 (Sampled Further)	0.0001	0.0006
	Micro-curies/ft.^{3}	Micro-curies/ft.^{3}
Air from Room I	0.00002	0.00001
Air from Hoods 1, 2, 3	0.00002	0.00001

* The hood nomenclature is that of drawing Number TD-782.

[redacted]

3.5 Particulate Discharge from Hot Hoods (Cont'd)

TABLE II

Building 205

Hood Stacks	Maximum Activity Level Mc/Hr.	Average Activity Level Mc/Hr.
Room I	0.00002	0.00001
Room II*	0.00002	.00001
Room IV, Hood 3	0.00007(5)	.00006

Note that in the preceding table for Building 706-C the air from Room I shows an activity level very near to that of the air discharged by the stack for hoods 1, 2, and 3 in room I.

Regarding Building 205, a fact worthy of mention is that during the sampling periods the amount of hot material handled by the 205 laboratory was below normal.

All together, eight stacks were sampled, and assuming all these stacks discharged their average activity for one day, the γ activity discharged would be 0.091 millicuries per day.

3.6 Active Gas and Particulate Discharge from Pile and from Pile Filter House

Exact evaluation of the total activity discharge (particulate and decaying gases) from the pile and from the pile filter house is a difficult undertaking. Considerable effort has been expended along this line but the results are hardly conclusive. Various data indicate the particulate output through the filter house is essentially zero; thus a number defining the filter house efficiency possesses academic interest only.

The Health Physics Division has concluded in several reports that the installation of the pile filter house resulted in a material reduction in area contamination. The reader is referred to those reports for further information.

---

* Hood and room numbers are shown on Drawing Number TD-781

[redacted]

[redacted]

3.6 Active Gas and Particulate Discharge from Pile and from Pile Filter House (Cont'd)

Some but not all of the efforts expended in the subject investigation will be here reported. It should be stressed that the unreported results in no way refute the conclusions about the filter house.

3.6.1 Pile Active Gas Output

The filter house inlet and exit air streams were sampled through a number of CWS #6 filter papers mounted as multi-ply layers in series. The papers were counted and analysed radio-chemically by the Chemistry Division for some of the major rare gas fission product chains.

A diagram of the sampling units is shown in Figure 19. The sampling flow was 4.8 c.f.m. out of approximately 100,000 c.f.m. pile air. The first layers of the upstream and down-stream units were located 4.8 sec. and 46.3 sec. respectively from the pile exit (the filter house CWS layer is 30 sec. from the pile exit). Flight time through the units (first multi-layer to second multi-layer) was 10.9 sec. The duration of each run was 3 days.

Table III is a summary of the activity observed on the papers two hours after removal from the apparatus. The papers are numbered from left to right as shown in Figure 19.

TABLE III

BETA ACTIVITY ON CWS PAPER*

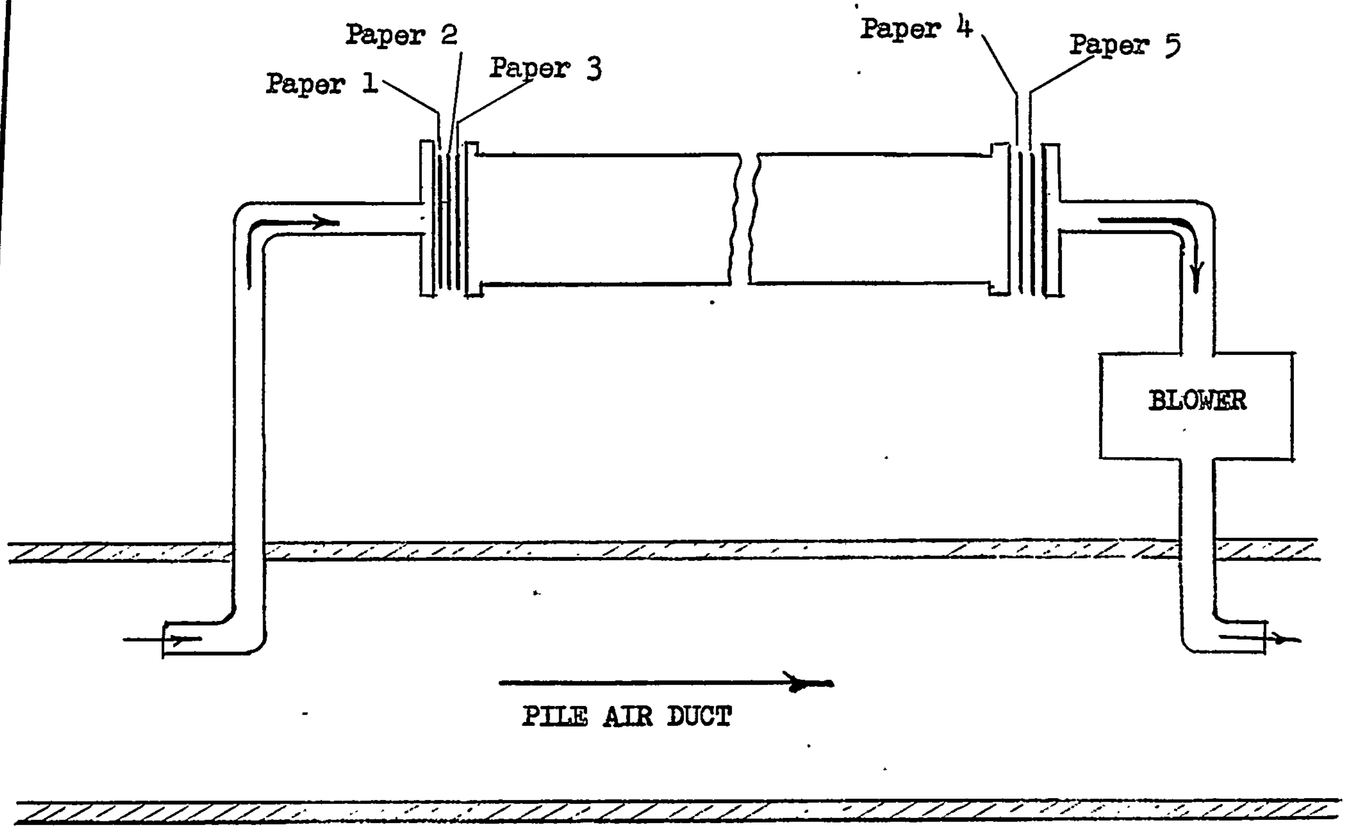
Second Shelf Counts Per Minute Per Sq. Inch.*

	House Inlet			House Exit		
Paper	Run 1	Run 2	Run 3	Run 1	Run 2	Run 3
1	29748	134367	43404	5478	4514	4021
2	508	796	579	1086	632	373
3	261	418	423	636	315	---
4	9555	8006	13996	2852	1265	3288
5	----	78	959	----	153	134

* Total area paper per filter 13.2 sq. inch.

[redacted]

FIGURE 19



FILTER HOUSE SAMPLING UNIT

[redacted]

[redacted]

3.6.1 Pile Active Gas Output (Cont'd)

No quantitative evaluation of the "particulate efficinecy" of the filter house was obtained from these data. Using mainly the radiochemical analyses it was possible to calculate the mean pile discharges of Kr 89, Kr 91, Xe 140, and Xe 141. The results of these calculations are given in Table IV.

TABLE IV

	Activity at Pile Exit Curies per day	Activity Test House CWS Layers Curies per day
Kr 89	12.5	12
Kr 91	200	34
Xe 140	45	19.5
Xe 141	150	0.2

The gaseous mothers remaining after the filter house give rise to active particulates. These, however, present no area contamination worry, amounting to but 5 - 10 millicuries per day of short-lived beta emitters and but about one millicurie per day of long-lived beta emitters. Further, particle sizes of the freshly formed material are within the Brownian range and are widely dispersed after exit from the stack.

That the filter house is effectively removing particulates (other than daughters of active gases) is supported by the fact that dust collections ahead of the filter house have yielded detectable quantities of uranium while uranium has never been detected in specimens obtained downstream of the filter house.

Further, the filter papers first in line after the filter house exhibited nothing but normal quantities of decay products (gaseous mothers)on analysis. Also the decay curves of down-stream papers agree rather well with the theoretical decays of the daughters (gaseous mothers) present at that point. The up stream papers show a decay much flatter than the theoretical daughter decay.

3.6.2 Radio-Autographs

The best evidence that the filter house is effectively removing gross particles is contained in the radio autographs,

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3.6.2 Radio-Autographs (Cont'd)

made by Mr. J. W. Gost, of filter specimens obtained during the work described in the preceding paragraph.

Figure 20 shows the radio-autographs of random specimens from a complete set of papers (upstream and downstream) taken over the same period. Gross particles (defined as those particles other than daughter particles) are characterized by the white specks. The haze effects are attributed to the decay-in-flight daughters of the active gaseous mothers.

3.7 Comparison of Active Particulate Sources

Table V is a convenient listing of the pertinent information for each potential source examined. Some data are included not reported previously.

4.0 ACKNOWLEDGEMENTS

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C. P. Coughlen

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April 24, 1950

Date

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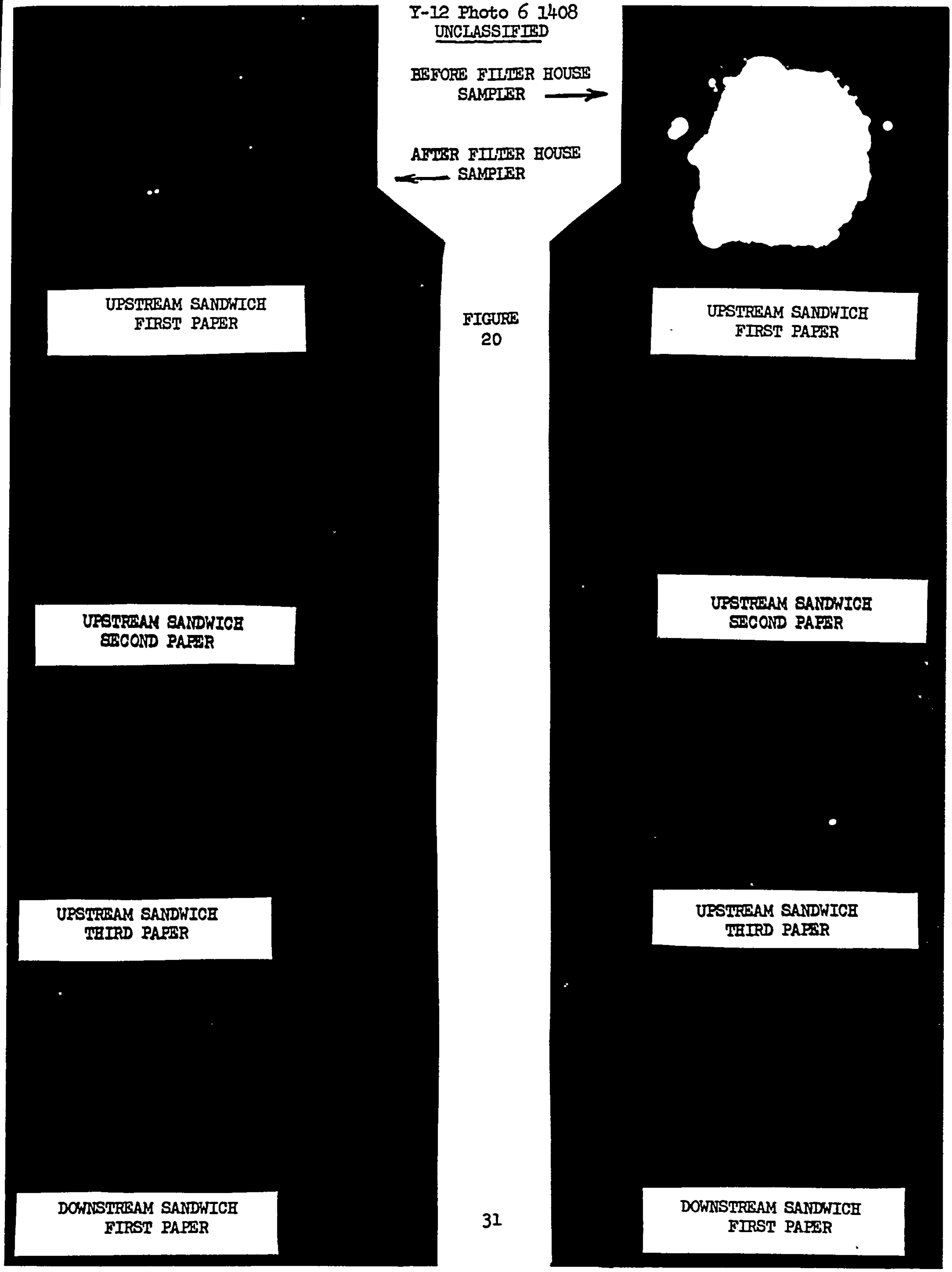

FIGURE 20

[redacted]

TABLE V

COMPARISON OF ACTIVE PARTICULATE SOURCES

Source	Air Flow CFM	Activity Concentration mc/ft³ 1 MEV/photon	Maximum Hourly Output mc 1 MEV/photon	Total Output mc/month 1 MEV/photon	Comments
Iodine-135 Operations					
Vessel Off-Gas	160	5.0×10^{-2}	1200	7600	Research operation now discontinued. Two runs per month. Mostly short-lived activity. One-day old slugs. Vessel line vented through 205 stack.
Cell Vent	3780	9.4×10^{-5}	118	340	
Total				7940	
RaLa Operations					
Cell Vent Before Filter	(2440)	(3.7×10^{-5})	(247)	(2710)	One run per month. Higher proportion of long-lived activity. Five-day old slugs. A-16 and A-4 lines are vented through 205 stack.
Cell Vent After Filter	2440	2.3×10^{-6}	4.2	165	
A-16 Line	80	1.1×10^{-3}	293	2700	
A-4 Line	30	2.4×10^{-3}	13.1	422	
Total After Filter				3300	
Redox Operations-Hot Pilot Plant					
30% Hanford Level					
Vessel Off-Gas	130	4.1×10^{-4}	7.0	1682	Operations now discontinued. Two runs per month. Long-lived activity only at 100% level. Discrepancy between 30% and 100% Hanford level is caused by 120-day aging at 100% level, and 40-day aging at 30% level. All Hot Pilot Plant lines are vented through 205 stack.
Dissolver Off-Gas	10	8.8×10^{-3}	8.0	424	
Cell Vent	20,000	8.5×10^{-8}	0.16	54	
			Total	2160	
100% Hanford Level					
Vessel Off-Gas	110	2.4×10^{-5}	0.483	120	
Dissolver Off-Gas	10	1.1×10^{-4}	0.067	4	
Cell Vent	20,000	3.0×10^{-8}	0.0848	30	
			Total	154	

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TABLE V (CONT'D)

COMPARISON OF ACTIVE PARTICULATE SOURCES

Source	Air Flow CFM	Activity Concentration mc/ft^3 1 MEV/photon	Maximum Hourly Output mc 1 MEV/photon	Total Output mc/month 1 MEV/photon	Comments
Pile Air-Before Filter	(100,000)	(3.6×10^{-8}*)	(5.0‡)	(154*)	*Based on total filter house and activity after 156 days of operation.
After Filters				1	‡Includes short-lived activity, but not active gases.
Iodine-131 Operations					
Cell Vent	4320	1.0×10^{-6}	0.54	75	Weekly runs. One day old slugs but interior scrubbers are quite effective.
Vessel Off-Gas	175	1.4×10^{-5}	0.49	42	Vessel line vented through 205 stack.
Total				117	
Hoods - 706-C	12,000	4.5×10^{-9}	0.01	2.3	Activity of room air close to hood stack gas activity.

NOTE: The total hood air discharge for ORNL has been estimated to be 215,000 cfm. If all the hoods were as hot as those in 706 C (doubtful), the total area contamination from this source would be 43 mc/month.

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